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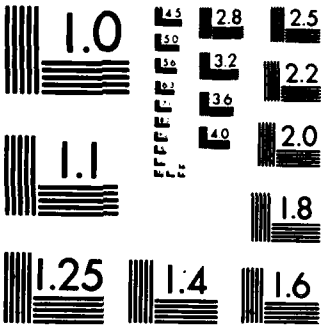

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Transport properties of very thin (2.5 monolayer) films of Ag epitaxially grown on clean Ge(001) substrates are reported. The films consist of a monolayer coverage plus isolated three-dimensional islands. Below ~70°K, the conductivity is dominated by the metal film and displays the temperature and electric and magnetic field dependences characteristic of metallic weak localization in two dimensions. Below ~2°K, the resistance drops rapidly in a manner resembling an incomplete superconducting transition. The resistance is restored by application of a magnetic field of ~20 KGauss @ 0.6

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TECHNICAL REPORT No. 3

ELECTRON LOCALIZATION AND SUPERCONDUCTIVITY OF VERY THIN EPITAXIALLY GROWN Ag FILMS ON Ge(001)

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ELECTRON LOCALIZATION AND SUPERCONDUCTIVITY IN VERY THIN EPITAXIALLY GROWN Ag FILMS ON Ge (001)*

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Transport properties of very thin (2.5 monolayer) films of Ag epitaxially grown on clean Ge (001) substrates are reported. The films consist of a monolayer coverage plus isolated three dimensional islands. Below ~70°K the conductivity is dominated by the metal film and displays the temperature and electric and magnetic field dependences characteristic of metallic weak localization in two dimensions. Below ~2°K the resistance drops rapidly in a manner resembling an incomplete superconducting transition. The resistance is restored by application of a magnetic field of ~20 KGauss at 0.6°K.

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In recent years there has been a renewed interest in the problem of two dimensional electronic systems with electron localization receiving a great deal of emphasis[1-23]. Single parameter scaling theories predict that all electron states are localized in two dimensions even when the conductivity is greater than the minimum metallic conductivity [R^{-1}_{2d}=(~30KΩ/square)$^{-1}$] predicted by Thouless[1].

In this communication we would like to describe a new metal film system which is ultra thin (down to approximately 2 Angstroms), crystalline and displays weak electron localization and behavior suggesting superconductivity at low temperatures. Some work on epitaxial thin metal films on semiconductors has been reported, but is somewhat complicated by compound formation[18,19].

The samples are prepared by epitaxial growth of the metal film on a semiconductor surface. Single crystals of germanium (Ge) the surface of which was oriented to within 0.5° of a (001) plane, were alumina polished and chemomechanically polished. The crystal was mounted in an ultra high vacuum chamber (base pressure <10^{-10} Torr). The surface was further cleaned in vacuo by several cycles of argon bombardment and annealing at ~825°K. The surface was structurally characterized using low energy electron diffraction (LEED) and Auger electron spectroscopy (AES). Silver (Ag) was then deposited using a pyrolitic graphite Knudsen effusion cell. The background pressure in the chamber was ~2x10^{-9} Torr during deposition. The Ag film grew epitaxially at a rate of about 2 Angstroms per minute. Deposition rate and Ag film thickness were measured using a quartz crystal microbalance (calibrated by using Rutherford backscattering (RBS)). LEED and AES were performed on the Ag film during growth. Details of the preparation and characterization of these films is more completely described in reference 24. LEED studies show that the Ag films grew by monolayer coverage followed by three dimensional island formation as deposition continued. Reference 25 on Ag on Ge (111) suggests strong chemical interaction and intermixing of the first several layers. However, other studies before[26] and since[24,27,28] reference 25 indicate that there is no interfacial alloying or interdiffusion of the Ag and Ge at room temperatures.

The samples were removed from the ultra high vacuum system and stored in air until they could be analyzed by scanning electron microscopy (SEM) or RBS. The samples were placed in a conventional evaporation system (base pressure 10^{-6} torr) and 3000 Angtrom Ag contacts were electron beam deposited onto the samples.

Electronic transport measurements were performed in a sealed, temperature controlled copper sample holder placed in an exchange gas can and submerged in liquid helium. Copper wires were indium soldered to the silver pads and heat sunk to the copper sample holder.

The monolayer film is certainly discontinuous, resting on plateaus on the Ge surface with step heights corresponding to 1/4th the lattice spacing of Ge. These plateaus are presumably bridged by the excess 3-Dim Ag islands, resulting in the observed conductivity dominated by the monolayers. The resistance of the sample at 10°K is ~7.7 kilo-ohms.

In figure 1 we have plotted the fractional change (relative to 10 Kelvin) in the 2-probe resistance of this sample as a function of the logarithm of the temperature. The exact geometry being probed is unknown due to the discontinuities discussed above. One can see however, that the sample resistance displays a logarithmically increasing resistance with decreasing temperature.

Figure 2 shows the fractional change in the dynamic resistance of this sample at various temperatures. At low fields the resistance behavior is consistant with a quadratic electric field dependence. At higher fields a logarithmic dependence appears which becomes more pronounced at lower temperatures. The insert shows the

change in the fractional resistivity with applied magnetic field. The magnetoresistance is isotropic indicating that orbital effects are negligable. This isotropic behavior is similar to that found in Pd films whose resistivities are above a few Kohms per square[19,20,22,23]

The previous figures show that these ultrathin crystalline films of Ag epitaxially grown on (001) Ge substrates exhibit electronic transport consistant with two dimensional behavior and display weak electron localization above approximately 2 degrees Kelvin. In figure 3 we show the resistivity of the film in figures 1-2 down to 0.65°K. The sample resistivity is increasing logarithmically with decreasing temperature until the temperature drops to about 1.6°K. Below this temperature the resistivity decreases rapidly. At 0.65°K its value is 55% of the resistivity at 2°K. A magnetic field suppresses this sudden decrease in resistance.

Heating the sample to ~600°K for ~10 min. under vacuum permanently destroys all signs of 2-d electronic behavior and superconductivity, consistant with the surface studies which indicate the monolayer should be unstable against island formation at that temperature[24].

In figure 4 we have plotted the perpendicular magnetic field behavior of the resistance of a similar sample whose resistance has been reduced (see insert) at 0.55°K to 15% of its 2°K value. Above ~1.4°K, the resistance increases immediately with the application of a magnetic field and then levels off at a value only slightly above the zero field resistance. At low temperature the magnetic field dependence is more dramatic. The resistance is flat for small fields, then increases abruptly and levels off at a resistance much higher than the zero field value.

Critical current measurements indicate a current density of 10^5-10^6 amps/cm^2 assuming the film is continuous and of uniform thickness.

We were concerned that exposure to air could damage the monolayer films. After these experiments were performed and repeated (over a period of approximately a month) the samples were reexamined by AES. This analysis showed that although the 3000 Angstrom Ag contact pads were tarnished (sulfur contamination), the only foreign substance on the 'monolayer' film was carbon - presumably from pump oil.

The scaling theory[3] predicts a logarithmically increasing resistivity with decreasing temperature. The prefactor of the logarithm depends on the exponent of the temperature dependence of the inelastic scattering time.

$$\frac{\Delta R(T)}{R} = -[Re^2 p/(2\pi^2 \hbar)] \ln(T/T_o) \qquad (1)$$

where the total inelastic scattering time τ_i goes as $\tau_i \propto T^{-p}$.

In between inelastic electron-phonon scatterings the electron can absorb energy from the applied electric field and undergo Joule (I^2R) heating. This results in a quadratic electric field dependence for low fields changing over at large fields to a logarithmic dependence. The prefactor depends on the power of the temperature dependence of both the total inelastic scattering time and the electron-phonon scattering time[5] ($\tau_{ep} \propto T^{-p'}$):

$$\frac{\Delta R(E)}{R} = -[Re^2/(2\pi^2 \hbar)][2p/(2+p')] \ln(E/E_o). \qquad (2)$$

An interacting electron picture[4,6,7] for a disordered system also suggests a logarithmic temperature dependence to the resistivity similar to equation (1) with p=1. Calculations in reference 13 indicate that aside from electron heating effects there is no electric field dependence to the resistivity in the interaction picture.

From our data we cannot determine the exponent p for the temperature dependence since the geometry is undefined as discussed above. Taking a value of p as 1 for example, we would find that the resistance of 7700 ohms is equivalent

to a measurement of ~5.5 squares in series. As one can see from equations (1) and (2), the ratio of the logarithmic slopes of the temperature and electric field dependences can be used to calculate the temperature dependence of the electron-phonon scattering time. Using the logarithmic electric field dependence of the sample resistance at 3°K one finds p' of equation (2) to give p'=2.6, which compares with P'=1.8 to 4.5 found for Pd films[22,23] and p'=1.3 reported for Pt films[21].

Both scaling theory and the interacting electron picture predict magnetoresistance effects in classically small fields ($\omega_c\tau<<1$) which are several orders of magnitude larger than the 'usual' positive magnetoresistance seen in metals. Electron orbital effects[6,8,15], Zeeman splitting[14,17], spin-orbit coupling[8,13,14,16], and impurity spin scattering can all make contributions to the magnetoresistance in both pictures. Zeeman splitting makes a contribution which while isotropic, manifests itself only in the interaction picture. There is a contribution from the Zeeman effect in the scaling theory only when spin-orbit coupling is included[14]. Spin-orbit coupling has a significant effect in both interaction and scaling pictures.

Considering that Ag and Pd are situated side by side in the periodic table this similarity in their magnetoresistive behavior is not suprising. The spin-orbit interactions couple as Z^4, where Z is the nuclear charge (Z_{Ag}=47, Z_{Pd}=46, so $\Delta Z^4/Z^4$ <10%).

The most striking feature of this work is the rapid decrease of the resistance below 2°K which we tentatively associate with an incomplete superconducting transition, as might be found in an inhomogeneous sample with a variety of T_c's. The magnetic field dependence shown in figure 4 is consistent with this interpretation if the 'critical field' for the highest T_c portion of the film is taken as indicative of Pauli limiting. A critical field of ~25KGauss is expected at low temperatures for a T_c of 1.6°K. The 'critial field' is roughly isotropic, again suggesting a spin pairbreaking.

While it is not clear what is responsible for the superconductivity, it should be noted that there are no known stable AgGe compounds. Ag_4Ge is metastable and superconducting at 0.85°K[29] and there are reports of quench condensed AgGe alloys made at 4.2°K which are superconducting at up to 1.6°K[30,31]. All of the surface characterization done on these samples indicates only weak interactions (on a chemical binding scale) between the monolayer Ag and the substrate, with no compound formation[24] and a sharp interface. Additional surface studies on this growth of Ag on Ge also suggest the absence of intermixing, chemical shifts or compound formation[27,28]. However, the resolution of the different surface probes cannot detect regions of the surface which comprise only several percent, and it is always possible, even if unlikely, that a small interconnected part of the surface may contain compounds that we cannot detect but which short out the rest of the surface as they go superconducting. Our critical current measurements would tend to argue against this.

If in fact the monolayer is superconducting, while bulk Ag is known not to be, there can be several explanations. Since the observed transition temperature is low, there is no need to invoke any mechanism other than the usual electron-phonon interaction and BCS superconductivity. A monolayer of Ag has a vastly different bandstructure and is interacting with quite different phonons than the bulk material. If further investigations point to another mechanism, it should be remembered that the monolayer metal film on a Ge substrate is an ideal system for studying the excitonic mechanism as proposed in reference 32, especially since references 27 and 28 indicate that there is negligible band bending

at the Ge-Ag interface.

In conclusion, we have shown that ultrathin crystalline films of Ag epitaxially grown on Ge (001) substrates exhibit electronic transport consistant with two dimensional behavior. They display weak electron localization, which crosses over to a superconducting regime below 1-2°K. At low temperatures, in the normal state, this system has a resistivity which increases logarithmically with decreasing temperature. At low electric fields the normal state resistivity is consistant with a quadratic field dependence changing over to a logarithmic dependence at high electric fields. There is a small positive isotropic magnetoresistance. Below approximately 2°K the resistivity displays a broad superconducting transition in both its temperature and magnetic field dependence.

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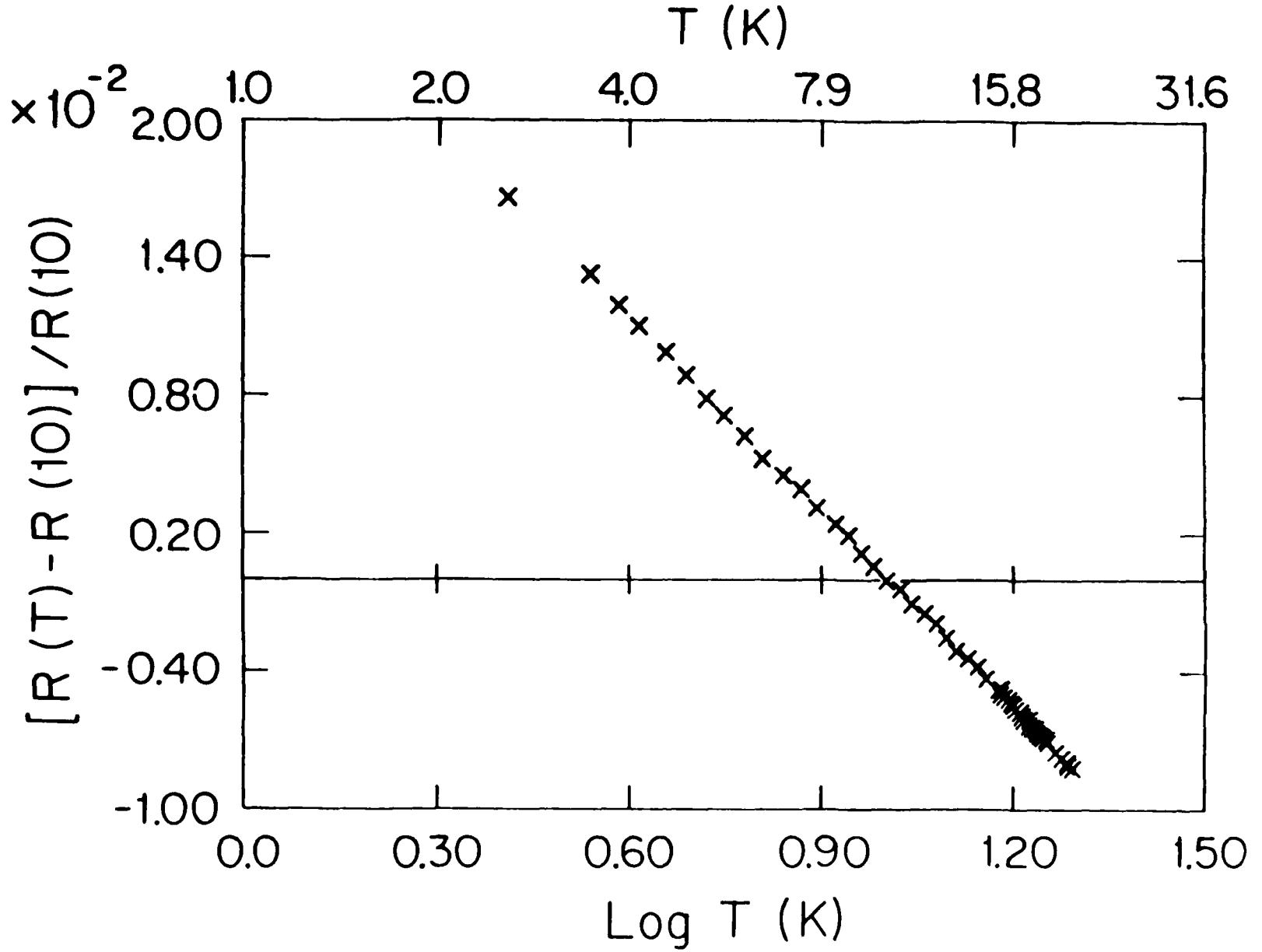
Figure 1
Fractional change in the film resistivity at low temperature. Microscopic terraces in the substrate prevent absolute resistivity measurements, but assuming p=1 in equation (1), would give a resistivity of ~1.4KΩ/square.

Figure 2
The dynamic resistance is consistant with a quadratic field dependence for low electric fields, changing to a logarithmic dependence at high fields. This sample gives p'=2.6 in equation 2. Insert shows a positive isotropic magnetoresistance, open symbols are for H ⊥ film, solid for H || film.

Figure 3
Below ~1.6°K the resistivity of this sample shows a very rapid decrease with decreasing temperature. The insert shows the sample resistivity from 250-0.6°K, which is well fit by a log(T) resistor in parallel with a semiconductor with a ~0.14eV gap.

Figure 4
The perpendicular magnetoresistance of a sample similar to that in figures 1-3, in fields up to 10 Tesla. Insert shows the temperature dependence of the resistivity below 25°K.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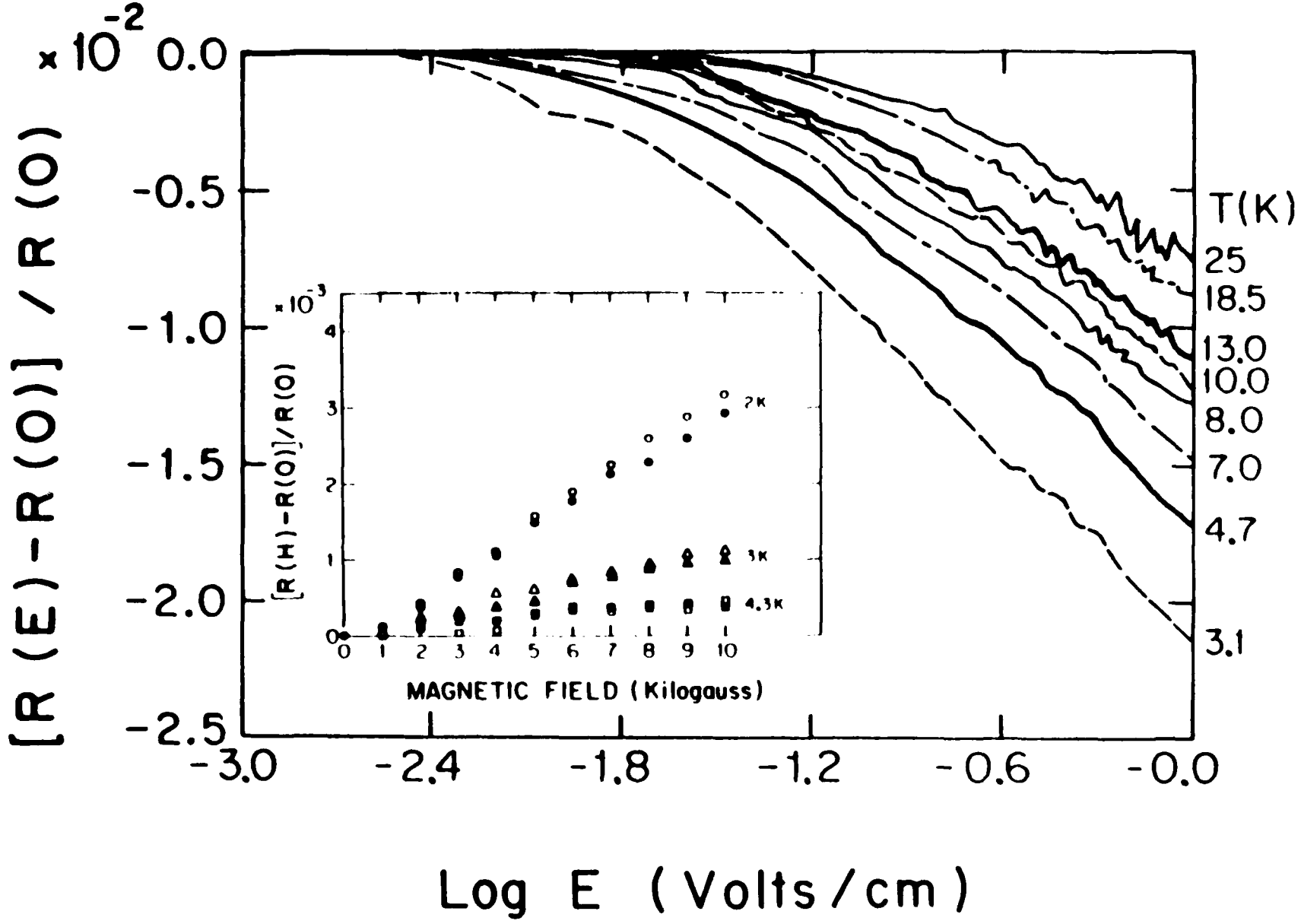
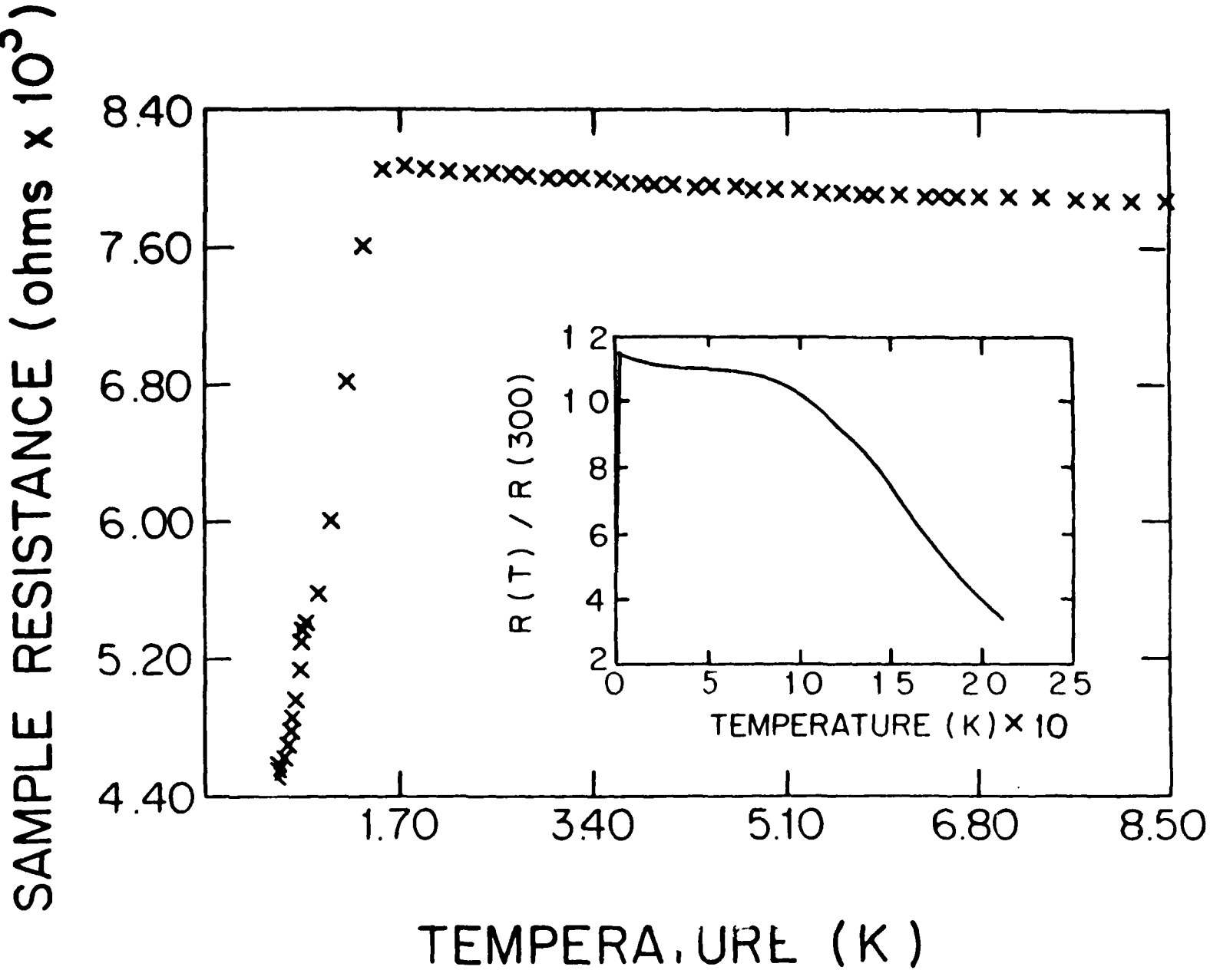
4.0
7.9
15.8
31.6
×10^-2
2.00
1.40
0.80
0.20
-0.40
-1.00
[R (T) - R (10)] / R(10)
0.0
0.30
0.60
0.90
1.20
1.50
Log T (K)


[R(E)-R(O)] / R(O)
x 10^-2
0.0
-0.5
-1.0
-1.5
-2.0
-2.5
-3.0
-2.4
-1.8
-1.2
-0.6
-0.0
Log E (Volts/cm)
T(K)
25
18.5
13.0
10.0
8.0
7.0
4.7
3.1
[R(H)-R(O)]/R(O)
x10^-3
2K
3K
4.3K
0 1 2 3 4 5 6 7 8 9 10
MAGNETIC FIELD (Kilogauss)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6.80
6.00
5.20
4.40
1.70
3.40
5.10
6.80
8.50
TEMPERATURE (K)
R(T) / R(300)
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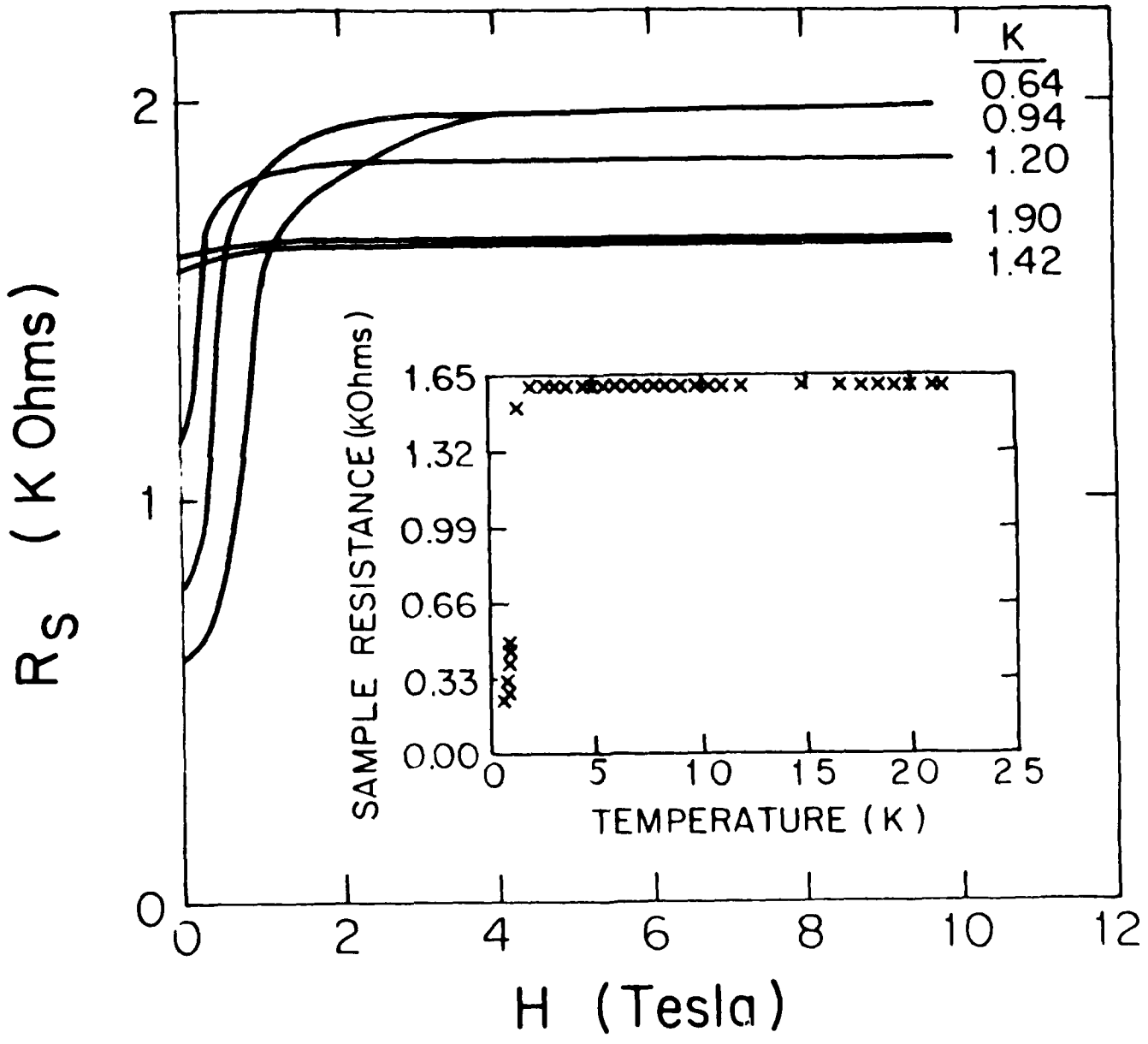
1.0
.8
.6
.4
.2
0
5
10
15
20
25
TEMPERATURE (K) x 10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1.20
1.90
1.42
R_S (K Ohms)
H (Tesla)
SAMPLE RESISTANCE (KOhms)
TEMPERATURE (K)
0
1
2
0
2
4
6
8
10
12
1.65
1.32
0.99
0.66
0.33
0.00
0
5
10
15
20
25